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"Unusual" temperature behavior of the photoluminescence of the InP and InGaAs quantum dots under quasiresonance excitation

V. Davydov†, *I. V. Ignatiev*†‡, I. E. Kozin†‡,
J.-S. Lee†, H.-W. Ren†, S. Sugou† and Y. Masumoto†¶
† Single Quantum Dot project, ERATO, JST, Japan
‡ Institute of Physics, St.-Petersburg State University, Russia
¶ Institute of Physics, University of Tsukuba, Japan

Abstract. A few "unusual" temperature effects are observed in the photoluminescence (PL) spectra and PL kinetics of heterostructures with self-assembled InP and InGaAs quantum dots (QDs) under the excitation within the PL band of QDs. It is found that the temperature rise leads to a *considerable increase* of the Stokes part of the PL. The analysis of this phenomenon and PL kinetics is based on the assumption about the slow relaxation of hot carriers.

Introduction

The usual method of PL excitation through the barrier interband transitions involves many relaxation processes for hot carriers. On the contrary, the excitation in the spectral region of the QD absorption creates the carriers inside QDs. Therefore, only a few relaxation processes occur prior to the radiative recombination of the electron-hole pairs. These processes can be studied in detail on different excitation conditions, at various temperatures, and under external electric field [ ], etc.

In this paper we study the temperature dependences of the QDs PL and also its kinetics under the resonant excitation of QDs. They reveal a "strange" behavior—apparent increase of the PL intensity with increased temperature. This behavior can be explained only assuming a slow energy relaxation of the photogenerated carriers.

1 Experimental

Several samples with InP and $In_{0.5}Ga_{0.5}As$ QDs were studied. They all reveal very similar temperature behavior of the PL. Here we show only a few examples of the experimental data. The samples with the $In_{0.5}Ga_{0.5}As$ QDs (No 140, No 141, and No 153) were grown by the MOVPE method on the semiinsulating (100) GaAs substrate. The QD layer is sandwiched by undoped GaAs epitaxial layers. The QD areal density was $\rho_{QD} = 5 \times 10^9\,cm^{-2}$ for No 140, $10^{10}\,cm^{-2}$ for No 141 and $2\times^9\,cm^{-2}$ for No 153. The structure with the InP QDs was grown by means of MOVPE (sample QDPG0298, $\rho_{QD} = 3 \times 10^9\,cm^{-2}$) and GS MBE (sample QDP1779, $\rho_{QD} = 10^{10}\,cm^{-2}$) on n^+ (100) GaAs substrates.

The PL spectra were measured by using a cw Ti:sapphire laser, a double monochromator U1000, and a photon counting system with cooled GaAs and InGaAs photomultiplier tubes. The sample temperature was varied using the Oxford gas-flow optical cryostat ("Optistat") in the range of 5–280 K.

The PL kinetic was studied using a picosecond Ti:sapphire laser (pulse duration 5 ps), a double monochromator with zero dispersion (spectral resolution 0.5 nm), and a streak camera. The time resolution of the setup was about 20 ps.

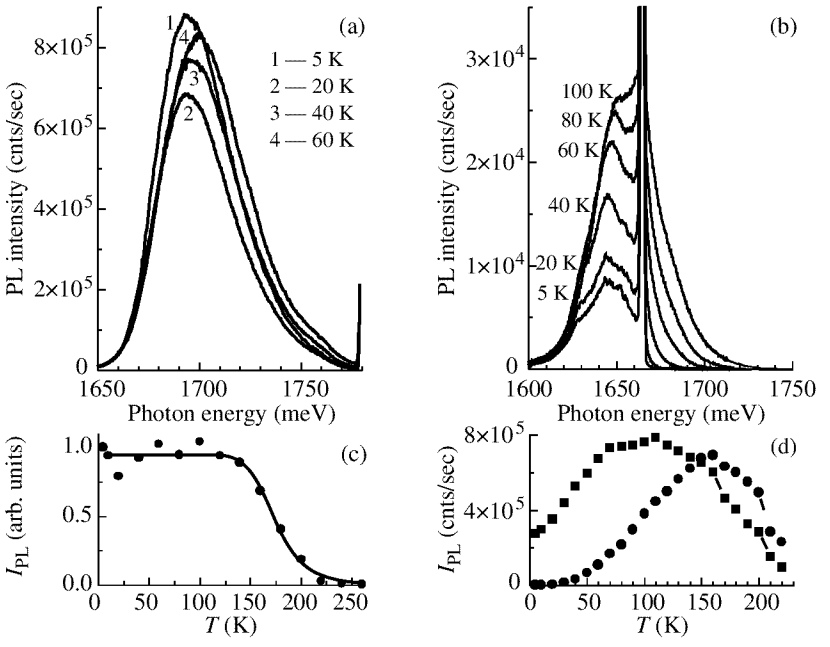

Fig. 1. General PL behavior of InP QDs at various temperatures. (a) PL under nonresonant excitation of $E_{exc} = 1784$ meV. (b) PL under resonant excitation of $E_{exc} = 1687$ meV. (c) $I_{PL}(T)$ under nonresonance excitation. Fitting by a model taking account of the activation energy type nonradiative process. (d) Dependence of integral Stokes (squares) and anti-Stokes (circle) PL intensity on temperature under resonance excitation.

2 Experimental results and analysis

The PL spectra of the sample QDPG0298 with InP QDs are shown in Fig. 1. Similar data for InGaAs QDs are presented in Fig. 2.

The main feature of the PL spectrum under resonance excitation at low temperature (Figs. 1(b) and 2(b)) is the presence of a dip between the laser line and the Stokes PL peak. This shows that the energy relaxation rate is small for small energy distance between levels in the QDs.

The temperature dependences of the integral PL intensity $I_{PL}(T)$ are different from each other for the nonresonance and resonance excitation (Fig. 1(c,d) and Fig. 2(b)). In the case of nonresonance excitation, a usual behavior with *decreasing* intensity at fairly high temperatures is observed. It can be explained by the process of thermostimulated quenching and described by the thermoactivation formula $I_{PL}(T) = I_{PL}(0)/[1 + a \cdot \exp(-\Delta E/kT)]$.

For the resonance excitation (Fig. 1(b,d) and Fig. 2(a)), *an increase* of the PL intensity is observed up to rather high temperatures. The integral intensity $I_{PL}(T)$ increases typically by 3–5 times for the most of the samples studied. We suppose that at low temperature a considerable fraction of the PL is resonant one and is covered by the exciting laser line. This is possible provided that the energy relaxation rate is comparable with or smaller than the radiative recombination rate. At elevated temperatures, the relaxation becomes faster due to the activation of the stimulated photon emission processes.

To prove this assumption, we have studied the time-resolved PL. The PL kinetics for the sample QDP1779 with the InP QDs are shown in Fig. 3. The PL intensity from a single

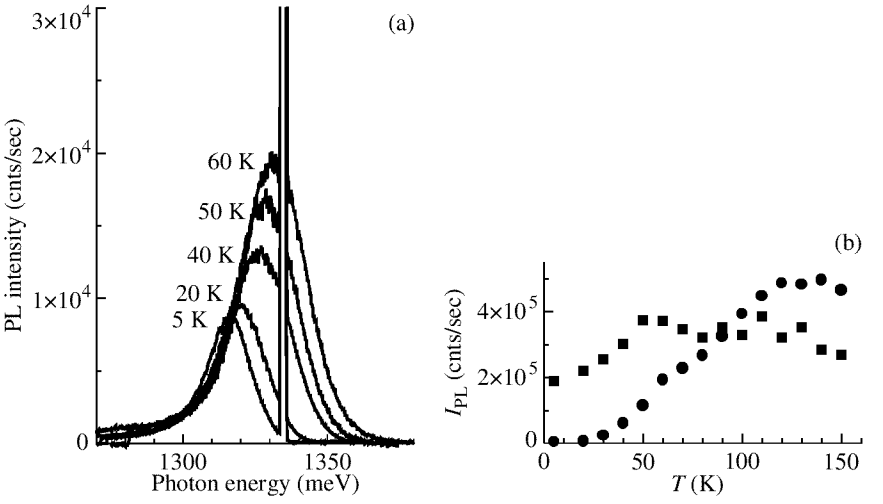

Fig. 2. General PL behavior with changing temperature for InGaAs QDs. (a) PL under resonant excitation $E_{\text{exc}} = 1334$ meV. (b) Dependence of integral Stokes and anti-Stokes PL intensity on temperature under resonance excitation ($E_{\text{exc}} = 1334$ meV).

layer of QDs is too weak compared with the background of the scattered laser light to detect directly the resonance PL. However, the spectral dependence of the kinetics reveals an increase of the decay time with increasing energy gap between the wavelengths of detection and excitation (Fig. 3(b)).

In our case, there is no grounds to anticipate energy transfer between the QDs, since the above effects are observed in structures with the areal densities of QDs varying by an order of magnitude. By this reason, the spectral diffusion can be related only to relaxation inside the QD.

As the temperature increases, the PL decay time τ becomes longer (Fig. 3(c)). This effect is usual for the system with continuous spectrum of energy states [2]. In the case of QDs, with a discrete spectrum of energy states, it can be explained by thermally activated population of nonradiative states. As the theoretical calculations [3, 4] show these states can be formed by the hole localization in small part of QDs. The temperature dependence $\tau(T)$ can be approximated in this case by the formula $\tau(T) = \tau(0)/(1 - ae^{-\Delta E/kT})$ which fits well the experimental data.

Under resonance excitation, increase of the temperature gives rise to the PL in the short-wavelength part of the spectrum above the exciting line (anti-Stokes PL, AS PL). Its behavior is rather different for the InP and InGaAs QDs and cannot be explained by taking into account only the Boltzmann statistics and the density of states extracted from the PLE spectra. We think that the AS PL profile is affected also by spectral distribution of the radiative recombination rate.

3 Conclusion

The above experimental data show that the behavior of the QDs ensemble is very similar in some aspects to behavior of a quantum-mechanical system with continuous energy spectrum. However, the physical origin of such a behavior has different nature and is related to slow relaxation of hot carriers, presumably due to their strong localization. There is no evidence for energy transfer between the QDs up to the temperatures when the PL

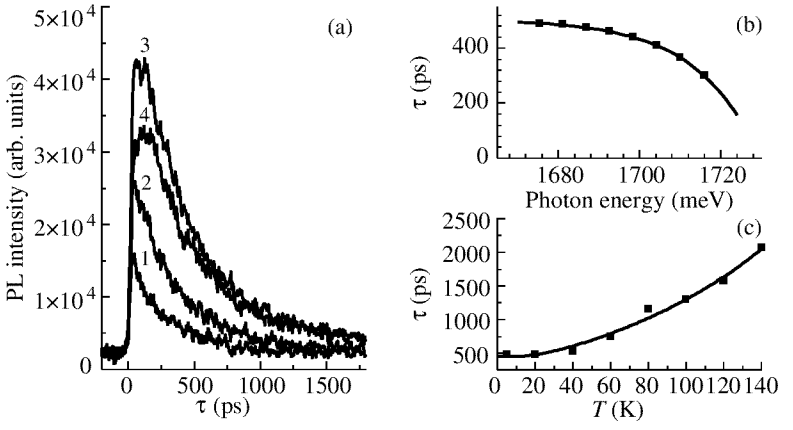

Fig. 3. (a) PL kinetics curve for the various detection energies. $E_{exc} = 1724$ meV, $E_{PL} = 1716.3$ meV for curve 1, 1710.3 meV—2, 1692.8 meV—3, 1681.4 meV—4. (b) Dependence of the τ on the detection energy at low temperature ($T = 5$ K). (c) Dependence of the τ of the PL at $E_{PL} = 1687.1$ meV on the temperature.

quenching starts.

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